Phase One Progress Report

INERT-MATRIX FUEL:
ACTINIDE "BURNING" AND DIRECT DISPOSAL

Nuclear Engineering Education Research Program
(grant # DE-FG07-99ID13767)

Rodney C. Ewing (co-PI)
Lumin Wang (co-PI)

Department of Nuclear Engineering and Radiological Sciences
University of Michigan
Ann Arbor, MI 48109

Reporting Period: 07/01/1999 to 06/30/2000

1. Background

Excess actinides result from the dismantlement of nuclear weapons (^{239}Pu) and the reprocessing of commercial spent nuclear fuel (mainly ^{241}Am, ^{244}Cm and ^{237}Np). In Europe, Canada and Japan studies have determined much improved efficiencies for burn-up of actinides using inert-matrix fuels. This innovative approach also considers the properties of the inert-matrix fuel as a nuclear waste form for direct disposal after one-cycle of burn-up. Direct disposal can considerably reduce cost, processing requirements, and radiation exposure to workers.

2. Objectives

The objectives of this NEER program are to study the nuclear fuel and waste form properties of the most promising inert-matrix fuels (i.e., cubic zirconia and spinel-zirconia composites). The three topics of study as listed in the original proposal are:

a) The effect of fission event (in-reactor) and alpha-decay event (in-repository) damage on the physical and chemical properties that are important to nuclear fuel (e.g., bubble formation and swelling) and nuclear waste forms (e.g., leach rate and micro-fracturing) performance.

b) The solubility and the mobility of the fission and other important transmutation products in the inert fuel matrix at both high (reactor fuel condition) and low (waste form condition) temperatures.

c) The determination of corrosion rate of ZrO_2 under expected repository conditions over long periods of time.

We have also stated in the original proposal that this research program would be conducted with international collaborations.

3. Research progress and accomplishments

Significant progress has been made during the first year of the research program on proposed topics a) and b) (see details in 3.1 and 3.2), and an unique leaching equipment has been ordered for starting the work on topic c). International collaboration with European Commission Joint Research Center in Germany has been initiated. A full time graduate student research assistant and a post-doctoral fellow have been hired and supported by this program. We are active participants in the International Inert Matrix Fuel Consortium.

3.1 Main results

To study the effects of fission of fission product incorporation and associated radiation damage in cubic zirconia that is considered as an inert nuclear fuel matrix, cesium, iodine and strontium ions have been implanted into yttria-stabilized cubic zirconia (YSZ) with ion accelerators at Michigan Ion Beam Laboratory or HVEM-Tandem DOE User Facility at Argonne National Laboratory. The ion implantation was initially conducted at room temperature to $1x10^{21}$ ions/m^2 for each ion with ion energies ranging from 70 to 400 keV. The peak displacement damage level and the peak ion concentration in YSZ reached 205-330 displacement per atom (dpa) and 11-26 at.%, respectively. The microstructure of the implanted YSZ was studied by *in situ* and cross-sectional transmission electron microscopy (TEM). In the iodine and strontium implanted samples, a damaged layer with a high density of defect clusters formed after the implantation, but amorphization did not occur. Strontium containing nanocrystalline precipitates were formed in the Sr-implanted specimen after annealing at 1000°C indicating the implanted strontium concentration has exceeded the solubility in YSZ. However, an amorphous layer in Cs-implanted YSZ was observed in areas with Cs-concentrations greater than 8 at. % which is the first evidence of solid state amorphization of cubic zirconia. Amorphization of YSZ is caused by a large size incompatibility and the low mobility of cesium ions in the cubic zirconia structure (reflecting the low solubility of Cs in zirconia) rather than from purely radiation damage effects. Radiation-induced amorphization in zirconia with high impurity contents can cause important changes in the thermodynamic stability and chemical durability of the zirconia used as an intert-matrix fuel or as a nuclear waste form. However, the Cs-concentration at which amorphization of YSZ occurred in this study (~8 at. %) is above the value that may be reached in the inert fuel matrix or the waste form (i.e., ~5 at. % assuming a 30 at.% Pu loading).

Because of its relatively low thermal conductivity, YSZ is actually not very desirable for use as an inert fuel matrix, but the Europeans have recently proposed a promising concept for the inert matrix fuel, that is to use a "hybrid" material in which actinide containing particles such as yttria-stabilized cubic zirconia (YSZ) are dispersed in a thermally conducting matrix, such as spinel ($MgAl_2O_4$). In the development stage, Ce is used as a simulant for Am, as the two elements have similar chemical properties. Through collaboration with Institute for Transuranium Elements, European Commission Joint Research Center at Karlsruhe, Germany, we have started a study of the fission gas accumulation in such "hybrid" materials. In this study, prethinned CeO_2- or YSZ-containing spinel specimens were implanted with 200-400 keV Xe ions at 600°C using the IVEM-Tandem Facility at Argonne National Laboratory. *In situ* TEM was conducted during the implantation in order to follow the evolution of the microstructure. At a fluence between $2.4x10^{20}$ to $3x10^{20}$ ions/m^2 (up to 50 dpa and 4.7 at %), large Xe bubbles of 50-100 nm developed at the boundaries of the ceria or YSZ particles, while high density of dislocation loops (up to 8 nm in diameter) and much smaller bubbles (up to ~4 nm in diameter) formed in the spinel matrix. No large bubbles were observed at the boundaries between the spinel grains. These results suggest that the boundaries between spinel and oxide particles are preferred sites for fission gas accumulation.

Our research program is on schedule, and we expect to continue as planned. The first year's budget has been fully expended.

3.2 Publications and presentations

1. L.M. Wang, S.X. Wang and R.C. Ewing, Radiation effects in cesium ion implanted zirconolite and zirconia. Presented at the International Global'99 Conference (August 29 to September 3, 1999, Jackson Hole, Wyoming).
2. L.M. Wang, S.X. Wang and R.C. Ewing, Effects of Cesium Ion Implantation in Yttria Stabilized Cubic Zirconia. Presented at the 5th International Inert Matrix Fuel Workshop (Paris, France, October 22, 1999).
3. L.M. Wang, S.X. Wang and R.C. Ewing, Amorphization of cubic zirconia by cesium ion implantation, *Philosophical Magazine Letters* 80 (2000) 341-347.
4. L.M. Wang, S.X. Wang, S. Zhu, R.C. Ewing, Effects of fission product incorporation on the microstructure of cubic zirconia. Presented at the 102th Annual Meeting of the American Ceramic Society (April 30-May 2, 2000, St. Louis, MO), submitted to *Journal of Nuclear Materials* for publication.
5. L.M. Wang, S. Zhu, S.X. Wang, R. C. Ewing, N. Boucharat, A. Fernandez and Hj. Matzke, Effects of xenon ion implantation in spinel-ceria and spinel-cubic zirconia composites. Presented by R.C. Ewing at the 6th International Inert Matrix Fuel Workshop, European Material Research Society 2000 Spring Meeting (May 30 – June 2, 2000, Strasbourg, France), Submitted to *Progress in Nuclear Energy* for publication.
6. L.M. Wang, S.X. Wang, S. Zhu, R.C. Ewing, Effects of fission product accumulation in cubic zirconia. Accepted for presentation at the Topical Conference on Plutonium Futures — The Science (July 10-13, 2000, Santa Fe, New Mexico).
7. R.C. Ewing, L.M. Wang, S.X. Wang and S. Zhu, Fission product incorporation in cubic zirconia as inert matrix fuel. Accepted for presentation at the 24th International Symposium on the Scientific Basis for Nuclear Waste Management (August 27-31, 2000, Sydney, Australia).
8. L.M. Wang, S. Zhu, S.X. Wang and R.C. Ewing, Formation of nanocrystals in strontium ion implanted cubic zirconia. Accepted for presentation at the International Conference on Ion Beam Modification of Materials 2000 (September 3-8, 2000, Rio Grande do Sul, Brazil).